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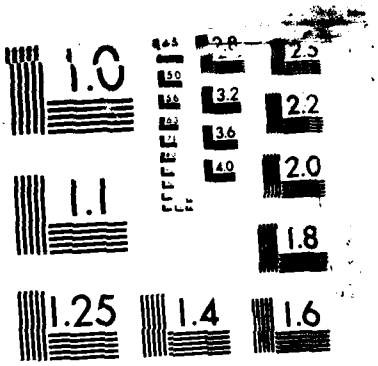

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NON-DESTRUCTIVE METHODS OF CHARACTERISING THE STRENGTH OF ADHESIVE-BONDED JOINTS

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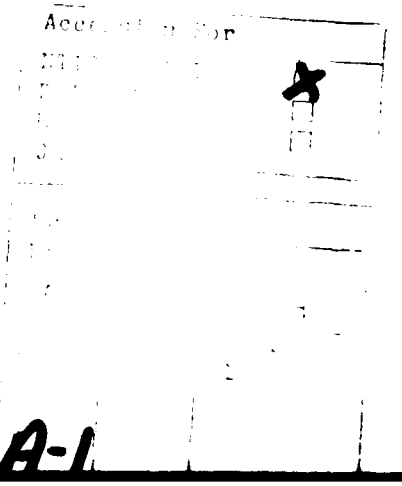
SUMMARY

Progress in the use of adhesive-bonded joints has been hampered by a lack of adequate non-destructive methods to check bond quality. This Memorandum describes briefly how this situation has arisen noting that, whilst many NDT methods give some measure of cohesive strength, it is adhesive strength that is of major concern. For joints with composite adherends it is concluded that it is surface contamination prior to bonding that must be sought, environmental degradation is not a major problem. With metallic adherends, however, the adhesive strength is strongly dependent on the detailed nature of the thin oxide layer and the way in which this becomes hydrated causing environmental degradation.

Ultrasonic methods are still considered to offer the best prospects but it is shown that sophisticated procedures will be necessary.



1 INTRODUCTION

Adhesive bonded joints can offer considerable advantages in terms of ease of construction and saving of weight. In recent years therefore there has been an increasingly widespread use of adhesive bonding in primary aircraft structure, but progress has been hampered by an inability to guarantee that the completed joint has an adequate strength. Rigorous process control and mechanical testing of representative samples has had to be employed in order to ensure that bond quality is maintained. Furthermore, environmental degradation can still give rise to concern.

Because of this, strenuous efforts have been made to develop non-destructive methods of interrogating the bond and detecting the presence of areas having inadequate strength. As a result a wide range of instruments has been developed but none of them provide a really satisfactory answer. Areas of disbond are generally readily detectable but, as will be shown in the next section, although some correlations of instrument response with bond strength have apparently been demonstrated such correlations are in fact of limited use.

RAE and Industry have had a number of discussions on this issue and this Memorandum is an attempt to summarise the current RAE view.

2 SOME BASIC CONSIDERATIONS

Before considering the possibility of non-destructive methods of evaluating bond strength it is necessary to draw two clear distinctions.

2.1 Adhesive strength and cohesive strength

The first is the distinction between the adhesive strength of the interface (or interfaces) between the adherend and the adhesive layer, and the cohesive strength of the adhesive layer itself. A reasonable estimate of the latter may be made by measurement of its elastic modulus, density and bond-line thickness. Although the relationship between these parameters and cohesive strength is somewhat empirical most of the existing NDT instruments (such as the Fokker Bond Tester) do in fact measure or respond to them in some way. Usually it is some parameter related to mechanical impedance or resonant frequency that is measured and it should therefore be noted that any variations in the mechanical properties of the adherend, as could happen when a composite is employed, will complicate the interpretation of such tests.

For reasons that will be explained shortly, measurement of the ahesive strength is a great deal more difficult. Indeed a recent authoritative textbook[1] concluded that "The cohesive strength of the adhesive is really the only parameter which can be estimated with any degree of confidence ...".

Having said this, however, there now seems to be general agreement that cohesive strength is not a matter of primary concern. This comment is based on opinions expressed at various meetings at which there has been both industrial and university representation. These included a meeting organised by the MOD/SBAC NDE Research Advisory Group in April 1983 which was attended by specialists in both adhesion science and NDE. The fact is that current design stress levels in the bond-line are very low compared with the

strength that is normally attainable, and that which is demanded by the mechanical quality control tests. Thus, on the rare occasions when process control fails to guarantee adequate cohesive strength, tests on off-cut coupons or travellers will readily reveal that this has occurred. Furthermore it is unlikely that the cohesive strength could be reduced to an unacceptable level without there being a marked change in the mechanical properties of the layer. Such a change would readily be revealed by conventional NDT techniques for, although they are primarily designed to look for disbonds, they will also respond to major defects such as gross porosity.

It is our opinion therefore that attention should be concentrated upon the possibility of developing methods to characterise the interface and hence hopefully to predict the adhesive strength.

2.2 The difference between metallic and composite adherends

It is at this stage that we need to draw the second distinction, which is that between metallic and composite adherends. For the purposes of this Memorandum consideration of composites will be limited to carbon fibre composites (CFC) having epoxy-resin matrices. There is, however, no reason to suppose that the conclusions reached are not equally applicable to aircraft-quality glass fibre composites, although the available evidence is more limited.

In broad terms the joint between a CFC adherend and an adhesive layer is a simple bond between two essentially similar materials. Furthermore, if such a joint is satisfactory immediately after fabrication then environmental degradation is not a major problem. This statement does, of course, assume that the reduction in strength (especially that at elevated temperatures) due to the uptake of moisture is taken into account in the design stress levels. To this must be added the caveat that there is at present very little known about any additional effects which may be introduced by fatigue loading, but the limited evidence available[2] suggests that fatigue loading at a realistic level has no effect on the residual strength.

In contrast the bond between a metallic adherend and the adhesive layer is an extremely complex one resulting from the procedures necessary to bond two very dissimilar materials. Environmental degradation is of course a major problem.

3 CFC ADHERENDS

Consider first the situation with a CFC adherend; the interface is not really a plane of weakness and a good joint will usually fail in the composite rather than the adhesive (unless a carrier is employed). The adhesive layer is no more sensitive to environmental degradation than is the resin matrix of the composite itself and there is no preferential degradation at the interface. The major concern is contamination of the surface of the composite prior to bonding and it is obviously far easier to detect such contamination at that stage. We are not in a position to state that such contamination could not be detected in a completed joint, but it would certainly be much more difficult. A possible area of concern on which we have no evidence could arise if, because of a geometric mismatch, the adhesive were to be cured with minimum or zero compaction pressure. It is possible to visualise a situation in which the adhesive was cured with no contact to

one adherend but in which intimate contact was established when the component cooled to room temperature. It seems unlikely, however, that these conditions would occur over significant areas; it is more likely that they would occur at isolated points and be accompanied by other areas of complete disbond which would, of course, be readily detectable. However, should such a situation occur, it would probably still be detectable but it would certainly require somewhat more sophisticated procedures than are currently applied. This could be an area where ultrasonic spectroscopy might prove helpful.

4 METALLIC ADHERENDS

With metallic adherends the situation is much more complex. In order to obtain a satisfactory bond it is necessary to prepare the metallic surface by etching and/or anodising to produce an oxide layer. Frequently a priming layer of some sort is also necessary in order to optimise the bond between the oxide and the adhesive. The adhesion scientists admit that their understanding of the processes involved is as yet far from complete[3]. It is not even certain how much of the strength is due to chemical bonding and how much to mechanical keying. There are in effect three separate interfaces which might need to be characterised:

(i) Metal to oxide
(ii) Oxide to primer
(iii) Primer to adhesive.

Of these the oxide-to-primer interface would appear to be most significant, but it should be remembered that failure can also occur within the oxide itself or within the primer. Attention has therefore been concentrated on characterisation of the oxide.

The properties of the oxide that are considered to be of importance are its thickness, its level of porosity and the degree of hydration. Other factors influencing the strength are the degree of penetration of the primer into the oxide pores and of course the presence of any contaminants.

An improved understanding of the bonding process is, of course, being actively pursued by the various adhesion groups and a wide range of laboratory techniques is being employed in an attempt adequately to characterise the oxide layer and the manner in which it is penetrated by the primer. It is to be hoped that in due course methods will be developed which will enable the surface pre-treatments to be monitored in a production environment. In the meantime, however, it would appear that there is no real substitute for strict process control.

A further difficulty arises with metallic adherends because, although a substandard surface pre-treatment can have an immediate effect on the as-fabricated strength, its more usual effect is to increase the vulnerability of the interface to environmental degradation. Thus poor pre-treatment will not necessarily be revealed by coupon testing at the time of manufacture.

It should be noted therefore that the primary task of an inspection of the pre-treated (and possibly primed) adherend is to detect those parameters which increase the susceptibility of the interface region to environmental degradation. With this in mind

it is clear that the non-destructive inspection of a completed joint is unlikely to be very informative with regard to the adhesive strength. If the subtleties of the interface cannot yet adequately be revealed by inspection prior to bonding then it is unreasonable to expect subsequent inspection to do so. Furthermore it is not even the current state of the complex and inaccessible interface that is required to be specified, it is those characteristics of the interface that will govern its future environmental performance.

5 IN-SERVICE INSPECTION

Attention has so far been concentrated upon the potential of non-destructive inspection during and immediately after fabrication. As noted earlier, with CFC adherends it is considered unlikely that there will be a requirement to monitor environmental degradation in service. Disbonds will, of course, require to be found in exactly the same way that delaminations are sought in the CFC. With metallic adherends, however, if the rate of degradation of the bond cannot be sufficiently well controlled by process control or predicted by inspection at the fabrication stage, then it may be necessary to seek some non-destructive means of monitoring this degradation.

What then, are we looking for as evidence that degradation is taking place? Environmental attack by water usually takes place at the adhesive (or primer)-oxide-metal interface. Stress corrosion of the metal substrate is not usually a major mechanism of environmental failure, although it is often a post-failure phenomenon. Kinlock[4] has stated that for aluminium alloys there is clear evidence that the locus of joint failure after environmental attack is through the oxide layer which has been weakened by the ingress of moisture. The weakened oxide is a hydrated form of the original oxide and the aim of an NDT technique would be to detect, and possibly to quantify, this oxide transformation. The evidence sought would probably be an increase in oxide thickness and a change in morphology; it is, however, possible that the hydration itself might be detectable by some means.

6 ULTRASONIC METHODS

The only NDT method that currently appears to have any real prospect of giving useful information on the state of the oxide in an assembled joint is some form of ultrasonic interrogation. Before briefly considering what might be achieved it may be helpful to note that the thickness of the oxide layer varies markedly with the pre-treatment process, but is always very much smaller than the wavelengths of the ultrasound.

Process	Average oxide thickness nm
Chromic-sulphuric pickle	40
Phosphoric acid anodise	400
Chromic acid anodise	2000
Wavelength of a 10 MHz compression wave in aluminium	600 μm (600000 nm)

Nearly all the attempts which have so far been made in the UK to monitor degradation by means of ultrasound have used a conventional pulse-echo approach in which the direction of the ultrasonic wave is normal to the plane of the adhesive layer. Electronic gating is used to isolate echoes from specific interfaces or sets of echoes from more than one interface. In order to identify the small changes in these echoes, which are very hard to distinguish in the conventional time-domain presentation, it has been necessary to process the signals and to present the data in the form of frequency spectra (and occasionally even cepstra).

Interpretation of the resultant spectra is, however, very difficult and various attempts have been made to model the propagation of ultrasonic waves in multilayer laminates; there has also been a good deal of supporting experimental work. Most of the models have, however, been highly idealised and have really so far only been of assistance in characterising the cohesive properties. Two convenient reviews of work in this area are available[5,6], and it is clear that strenuous efforts have been made to refine the techniques and to extract the maximum amount of information from them. So far, however, the only real success has been in obtaining correlation with cohesive properties. Indeed, in his revue[6] Curtis included the statement that "The technology required to examine the associated time and frequency domain exists but the 'Holy Grail' of adhesion strength still resolutely defies non-destructive evaluation by pulse-echo means".

Essentially there are three parameters which primarily affect the ultrasonic response in a simple pulse-echo system.

(i) The transit time of the ultrasonic pulse through each of the two adherends.

(ii) The transit time of the ultrasonic pulse through the adhesive layer.

(iii) The reflection (and transmission) coefficients at the adhesive-to-adherend interfaces.

Now the transit time through the adhesive layer is dependent on the velocity in that medium and on the thickness of the adhesive layer, both of which will be affected by moisture uptake by the adhesive. There will therefore be changes in the ultrasonic response due to the fact that the adhesive has taken up moisture but, although this may perhaps be related to cohesive strength, it gives no information on the condition of the oxide and hence on the adhesive strength. The fact that the initial thickness of the adhesive layer will in practice vary significantly from point to point also reduces the value of such information.

The reflection coefficient on the other hand is governed by the relative values of the acoustic impedance (the product of velocity and density) on either side of an interface between two materials. Now, as was shown above, the thickness of the oxide layer is much smaller than the wavelength of the interrogating ultrasound and it might at first sight be supposed that its effect would be negligible, and that the reflection coefficient at the complex adherend-to-adhesive interface would simply be governed by the impedances of the two major components. To some extent this is true and, since the reflection

coefficient is governed by the acoustic impedance (and hence by the elastic properties) of the adhesive, it too will be affected by moisture uptake in the adhesive. This latter effect can, in fact, cause confusion by being interpreted as a measure of the interface condition.

The presence of a thin intermediate layer, such as an oxide film, will effectively provide a frequency dependent modulation of the fundamental reflection coefficient. The magnitude of this effect is dependent upon both the thickness of the layer and its acoustic impedance. The better the acoustic match between the oxide and the aluminium adherend the less will be the degree of modulation. No data appears to be available on the acoustic impedance of a representative oxide but, since text-book values for an unspecified form of aluminium oxide are nearly twice that of aluminium, one is led to wonder whether a porous oxide has an impedance which differs very much from that of the parent sheet. If that were the case then it would not be detectable.

However, even if the presence of the oxide does provide a significant degree of modulation it must be remembered that it will probably be revealed as a somewhat minor change in a complex spectrum which will in any case be modified by environmentally induced changes in the adhesive itself. It is not surprising therefore that changes in the oxide condition have not so far been able to be identified.

Much of the pioneering work in this area was done at City University under MOD funding and some of this work is reported in Ref 7. A more complete description is contained in the final report on the contract[8]. These early results appeared promising and an attempt was made[9] to employ similar techniques in an industrial laboratory where more representative bonded components could be produced. Despite considerable care, however, it has not yet been possible to demonstrate a satisfactory relationship between variations in pre-treatment procedures and the resultant spectra.

Mention should be made of alternative methods of ultrasonic interrogration in which waves propagating parallel to the boundary surfaces are employed. These have recently been reviewed by Pilarski[10]. It would appear that the velocity or dispersion characteristics of surface or interface waves might give useful information on the state of an adhesive-to-metal interface, but the bonds employed in this limited experimental programme were unrepresentative of those encountered in aircraft structures. This approach should certainly not be dismissed, however, because it does appear to have rather more potential then the pulse-echo method with regard to characterisation of the oxide layer. Some supporting evidence for this was provided by Claus and Kline[11] who bonded borosilicate crown glass to Pyrex using an anaerobic cement. A range of surface finishes was introduced into the crown glass specimens prior to bonding by polishing the surfaces with different grades of carborundum optical abrasive. Measurement of the attenuation of Stoneley (interface) waves readily revealed the presence of the different surface finishes. They suggested that similar methods might be used to assess the effects of chemical contamination but no further publications are reported in Pilarski's review.

At present therefore it can only be stated that, although interface waves would be more difficult to generate, they may be able to provide more information about the state

of the oxide-to-primer interface for a fully cured adhesive. Once again, however, any changes in the bulk properties of the adhesive layer will have a strong effect and could well mask any effects due to changes in the oxide layer.

7 CONCLUSIONS

(1) It is adhesive rather than cohesive strength which is the matter of primary concern.

(2) With carbon fibre composite adherends the main concern is contamination of the surface prior to bonding. If such a joint is satisfactory immediately after fabrication then environmental degradation is unlikely to be a major problem.

(3) With metallic adherends it is the nature of the oxide produced by the etching and/or anodising process that is of prime concern.

(4) Improved characterisation of this oxide is required prior to the assembly of the joint.

(5) Environmental degradation with metallic adherends is through hydration of the oxide and will probably be revealed by a change in the thickness and morphology of the oxide layer.

(6) Ultrasonic interrogation currently appears to offer the best prospects for in-service inspection, although there is as yet no adequately developed method.

Acknowledgments

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17. Abstract

Progress in the use of adhesive-bonded joints has been hampered by a lack of adequate non-destructive methods to check bond quality. This Memorandum describes briefly how this situation has arisen noting that, whilst many NDT methods give some measure of cohesive strength, it is adhesive strength that is of major concern. For joints with composite adherends it is concluded that it is surface contamination prior to bonding that must be sought, environmental degradation is not a major problem. With metallic adherends, however, the adhesive strength is strongly dependent on the detailed nature of the thin oxide layer and the way in which this becomes hydrated causing environmental degradation.

Ultrasonic methods are still considered to offer the best prospects but it is shown that sophisticated procedures will be necessary.

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